EFFECT OF URANIUM SOLUBILITY ON RELEASE RATES FROM THE PROPOSED YUCCA MOUNTAIN REPOSITORY

E. E. Morris, T. H. Fanning, and R. A. Wigeland
Reactor Analysis and Engineering Division
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

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ABSTRACT

The sensitivity of the release of radionuclides from the engineered barrier system in the proposed Yucca Mountain Repository to the solubility of uranium is investigated. Factors ranging from 0.1 up to 100 were applied to the nominal uranium solubility assumed in one of the TSPA models used in support of the site recommendation for the repository. At times earlier than about 50,000 years, the release rate of uranium is proportional to the change in the solubility. By 100,000 years, the proportionality continues to hold when the solubility is reduced, but when the solubility is increased, the release rate changes by a factor less than the factor applied to the solubility. At times beyond about 300,000 years, when the solubility is varied from 0.1 to 100 times its nominal value, the release rate changes by less than a factor of 20. Over the same range of changes in the uranium solubility, changes in the release rates of uranium decay products are less than a factor of three. Because uranium and its decay products make relatively small contributions to the dose rate, the changes in the dose rate at a well located 20 km from the repository are estimated to be less than 20%.

INTRODUCTION

This paper describes a series of calculations investigating the effect of uranium solubility in water on the release from the engineered barrier system (EBS) of the proposed Yucca Mountain Repository. One of the total system performance assessment (TSPA) models[1] developed by the Yucca Mountain Project (YMP) in support of the site recommendation is used for the calculations. In the TSPA model, nearly 90% of the waste packages are placed in environmental conditions characterized by the absence of dripping water. However, the model assumes that there is a continuous film of water in the porosity of the waste package contents and in the porosity of the supporting structural material in the repository drifts so that diffusive transport is possible once the package is breached. The calculations described below show that when the uranium solubility is increased, the importance of diffusive transport to the release of uranium from the EBS also increases.

One of the key parameters estimated by the TSPA model is the dose rate received by individuals using water from a well located 20 km from the repository. Waste packages in the repository begin to fail after a few ten thousand years. Prior to about 50,000 years, the dose rate is due primarily to fission products, principally ^{99}Tc. After about 60,000 years, the main contributor to the dose rate is ^{237}Np. The peak dose rate occurs at about 270,000 years. During the time period beyond a few hundred thousand years, several uranium decay products make small but significant contributions to the dose rate. Among these are isotopes of protactinium, thorium, actinium, radium, and lead. In the TSPA geosphere model, the model describing water movement in the unsaturated and saturated zones in the repository, these elements are more strongly sorbing than uranium. Their retarded movement through the geosphere and their short half-lives (short compared to uranium) mean that, in some cases, their contribution to the dose rate depends primarily on the decay of uranium in the vicinity of the well. Increases in the solubility of uranium may increase the importance of some uranium decay products as contributors to the dose rate. Estimates of the impact of uranium solubility on the dose rate are made in the next to last section.

TSPA MODEL AND MODIFICATIONS

The starting point for the investigation is the GoldSim[2] model file SR00_042nm6.gsm, developed by the Yucca Mountain Project. This model produced the plot of the dose rate at the 20-km well over a 1,000,000-year time period shown as Fig. 4-188 in the *Yucca Mountain Science and Engineering Report*.[1] Within the model, all waste packages are placed in five infiltration bins. Packages within a bin experience the same infiltration rate. In each bin, the packages are

further divided into groups that always, intermittently, or never experience dripping water. Those packages that are never exposed to dripping water are, nevertheless, always wet.

Since the current study is focused primarily on release from the EBS, modifications were made to the GoldSim model file to shorten the time and reduce the memory required to perform calculations. First, transport of radionuclides through the unsaturated and saturated zones of the repository was removed from the model. It should be noted that the Yucca Mountain TSPA model does not impose solubility limits in the geosphere (unsaturated and saturated zones), so changes in the uranium solubility influence transport through the geosphere only indirectly through changes in the release rate from the EBS. Second, the need for performing time consuming calculations of time-dependent waste-package failure distributions and breach sizes on each run was eliminated. Elimination was accomplished by archiving failure and breach size distributions from one run and using the archived distributions on subsequent runs. These changes shortened the run time for individual time histories from more than one hour to between five and ten minutes without affecting numerical results.

Finally, a multiplier to be applied to the solubility of uranium in water was introduced. In the TSPA model, after a waste package fails and the waste form begins to degrade, radionuclides first enter a mixing cell called the waste form cell. When they leave the waste form cell, they pass through a second mixing cell representing the invert (the structural material placed in the bottom of emplacement drifts to support waste packages and waste handling equipment). The multiplier was used in the waste form and invert cells for each infiltration bin and each dripping condition within an infiltration bin. When radionuclides leave the invert cell, they enter the collector cell that represents the boundary of the EBS. High outflows assigned in the collector cell make the radionuclide concentration in this cell essentially zero. When the multiplier was set to unity, verification calculations demonstrated that EBS release rates evaluated with the modified TSPA model were identical with those found in the original model file.

RESULTS

Impact of Solubility on Uranium Release

Figure 1 shows the release rate of ^{235}U from the

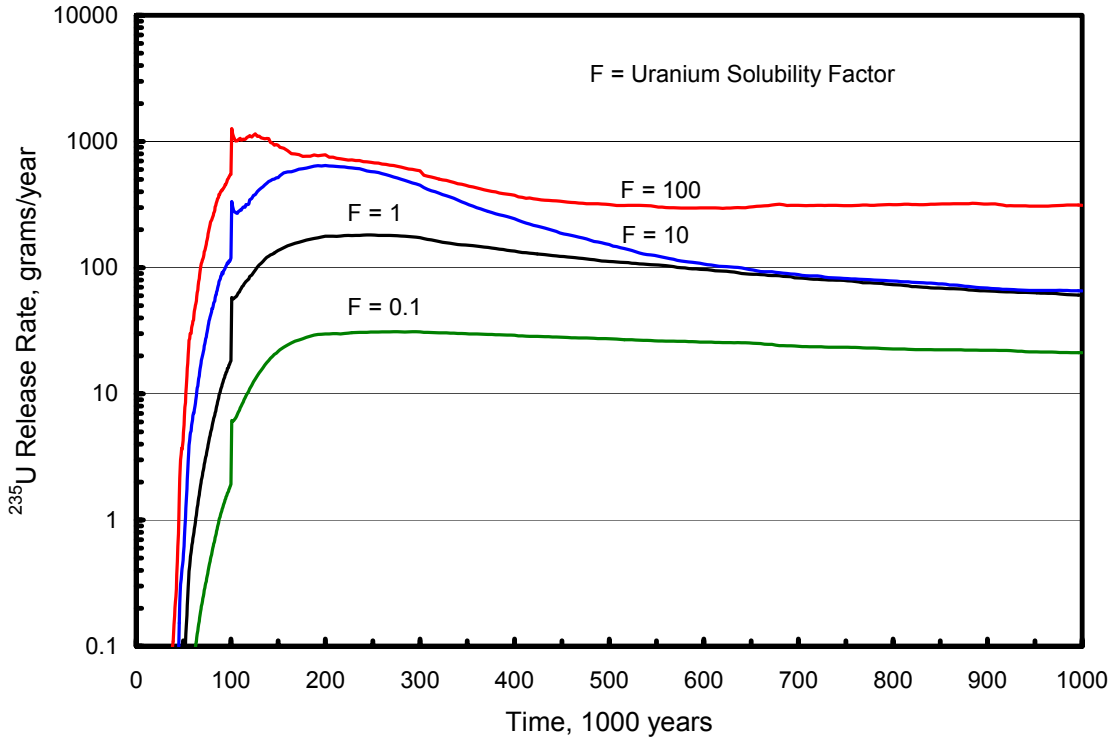

Fig. 1. Release rate of ^{235}U from the EBS as a function of the factor applied to the uranium solubility.

EBS for the nominal uranium solubility and for cases in which the solubility is decreased by a factor of 10 and increased by factors of 10 and 100. As for all the results reported here, the curves in Fig. 1 are the mean values of time histories for each of 300 independently sampled sets of the input parameters. At times around 50,000 years, the release rates change by essentially the same factor as was applied to the uranium solubility, indicating that the uranium release is solubility limited at this time. However, even as early as 100,000 years, the changes in the release rates are no longer proportional to the change in the uranium solubility. For example, at 100,000 years, reducing the uranium solubility from its nominal value to one-tenth its nominal value decreases the release rate by a factor of 10, indicating that the release is solubility limited. But increasing the uranium solubility by a factor of 10 causes an increase in the release rate of about a factor of six. Increasing the uranium solubility by another factor of 10 results in an additional increase of less than a factor of five. To understand this behavior, it is necessary to look at the release rates from the individual source types and consider differences in the solubility impact on packages in dripping and non-dripping conditions.

In Fig. 2, the ^{235}U release rates corresponding to the nominal uranium solubility are shown for both commercial spent nuclear fuel (CSNF) and co-disposal (CDSP) waste packages. These curves show that prior to 100,000 years, the release rate is dominated by the CDSP waste packages. Between 100,000 and 200,000 years, a transition occurs. For most of the time period after 200,000 years, CSNF waste packages are the largest contributor to the release rate. CDSP waste packages contain both DOE spent nuclear fuel (DSNF) and defense high-level waste glass (DHLW). Both these waste forms release their entire inventory within a few ten thousand years following waste package failure. Since all waste packages fail within a few hundred thousand years, it is the uranium solubility that prevents the release rate from the CDSP waste packages from dropping rapidly to zero after a few hundred thousand years. As solubility increases from the nominal value, peak value of the release rate for CDSP waste packages increases, but as time approaches 1,000,000 years the release rate decreases. When the uranium solubility is decreased from the nominal value, both the peak release rate and the release rate near 1,000,000 years decrease for CDSP waste packages. Release rates from CSNF waste packages increase or decrease with increasing or decreasing solubility at all times.

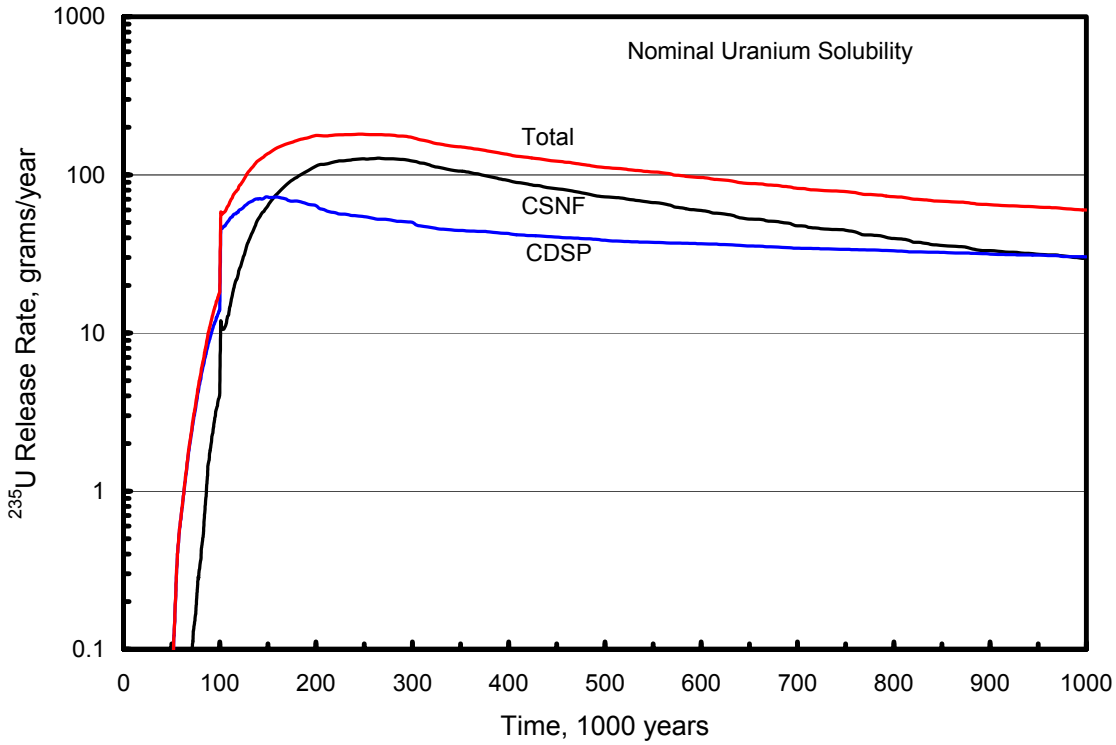

Fig. 2. Release rate of ^{235}U from the EBS for commercial and co-disposal waste packages with the nominal uranium solubility.

As described earlier, radionuclides released from the EBS pass from the mixing cell representing the invert to a collector cell. In the unmodified TSPA model, this collector cell serves as the source term for radionuclide transport through the geosphere. Because no solubility limits are imposed in the collector cell and outflows are assigned very high values, the radionuclide concentration in the collector cell is essentially zero. Since diffusive transport is driven by a concentration gradient, as long as the concentration in the invert cell is at the solubility limit, the high collector cell outflows will cause the gradient to be proportional to solubility.

Importance of Advective vs. Diffusive Transport

Figure 3 shows ^{235}U release rates for CDSP waste packages in infiltration bin 5. This bin contains the waste packages experiencing the highest infiltration rates. The curves show release rates for waste packages that either always, intermittently, or never experience dripping water. On average, 87% of the waste packages are placed in locations that never experience dripping water.[3] As noted earlier, the TSPA model assumes that a film of water, assumed present in the porosity of materials within these waste packages and in the invert below the packages, provides a medium through which radionuclides can move by diffusion. For waste packages that always or intermittently experience dripping, advective transport is the dominant transport mechanism. Because the waste forms in co-disposal waste packages degrade rapidly, the release rate of uranium from packages in always or intermittent drip environments show high peak values. These high values deplete the uranium inventory quickly and the release rate declines rapidly with increasing time. As the uranium solubility is increased, diffusive transport from waste packages in non-dripping environments becomes more important. As long as there is a sufficient amount of uranium to keep the concentration within the waste package or the invert at the solubility limit, the concentration gradient for diffusive transport is proportional to the solubility. However, for CDSP waste packages, when the solubility is increased by a sufficient amount, even diffusive transport exhausts the uranium inventory. As a result, the release rate from waste packages that are never dripped upon increases at early times with increasing solubility but then declines more rapidly with increasing time as the uranium inventory is used up.

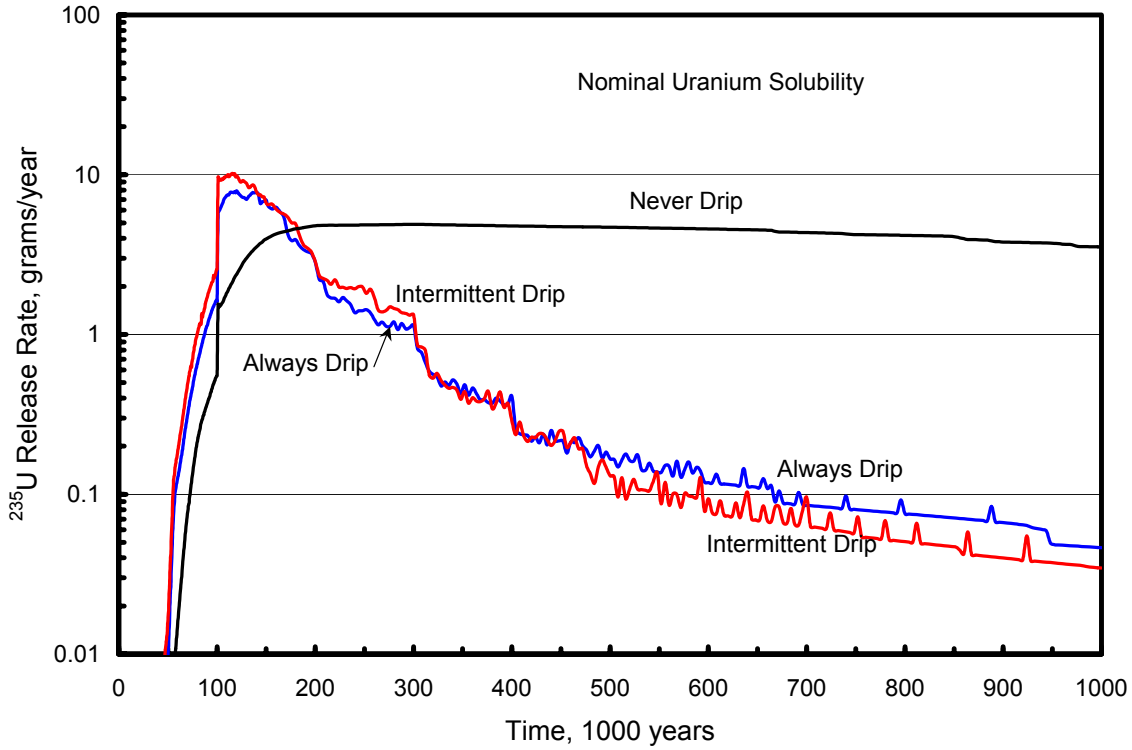

Fig. 3. Release rate of ^{235}U from the EBS for co-disposal waste packages in different the dripping environments of infiltration bin 5 with the nominal uranium solubility.

Similar factors influence the release rates of uranium from the CSNF waste packages, but the results do not look the same because of different waste form behavior. In addition, individual CSNF waste packages have about six times as much ^{235}U as CDSP waste packages and more than ten times as much ^{238}U. The larger inventory of uranium has the consequence that uranium concentrations stay at the solubility limit for longer times. Thus, when the uranium solubility changes from its nominal value, the release rate due to diffusive transport in never-drip waste packages changes accordingly. This pattern begins to break down when the uranium solubility is increased from 10 times nominal to 100 times nominal. Waste packages in always or intermittent drip environments are not as strongly influenced by the uranium solubility. The release rate decreases by about a factor of 10 when the uranium solubility decreases by a factor of 10. But when the uranium solubility is increased by a factor of 10 from the nominal value, release rates change by less than a factor of 10. And, when the solubility is changed from 10 times nominal to 100 times nominal, the release rates change very little. At the higher uranium solubilities, the rate of release of radionuclides from CSNF waste packages in always or intermittent drip environments is controlled by the degradation rate rather than the advective transport rate. The degradation rate for CSNF is significantly lower than for DSNF and DHLW.

Collector cells with high outflows are a computational convenience and cause the importance of diffusive transport for waste packages, especially in never drip environments, to be exaggerated. In reality, the concentrations of radionuclides leaving the invert probably would not drop abruptly to zero. In the event that this aspect of the TSPA model is changed, it may be desirable to reassess the results obtained here with the newer models.

Impact on the Release of Uranium Decay Products

The release rate for ^{231}Pa is shown in Fig. 4 as a function of the change in uranium solubility. ^{231}Pa is a daughter of ^{235}U and contributes a small but non-negligible amount to the dose rate at the 20-km well. Because there are relatively small amounts of ^{231}Pa in the waste, after a few hundred thousand years, the dominant source for this isotope will be the decay of ^{235}U. As shown in Fig. 1, the uranium isotopes are

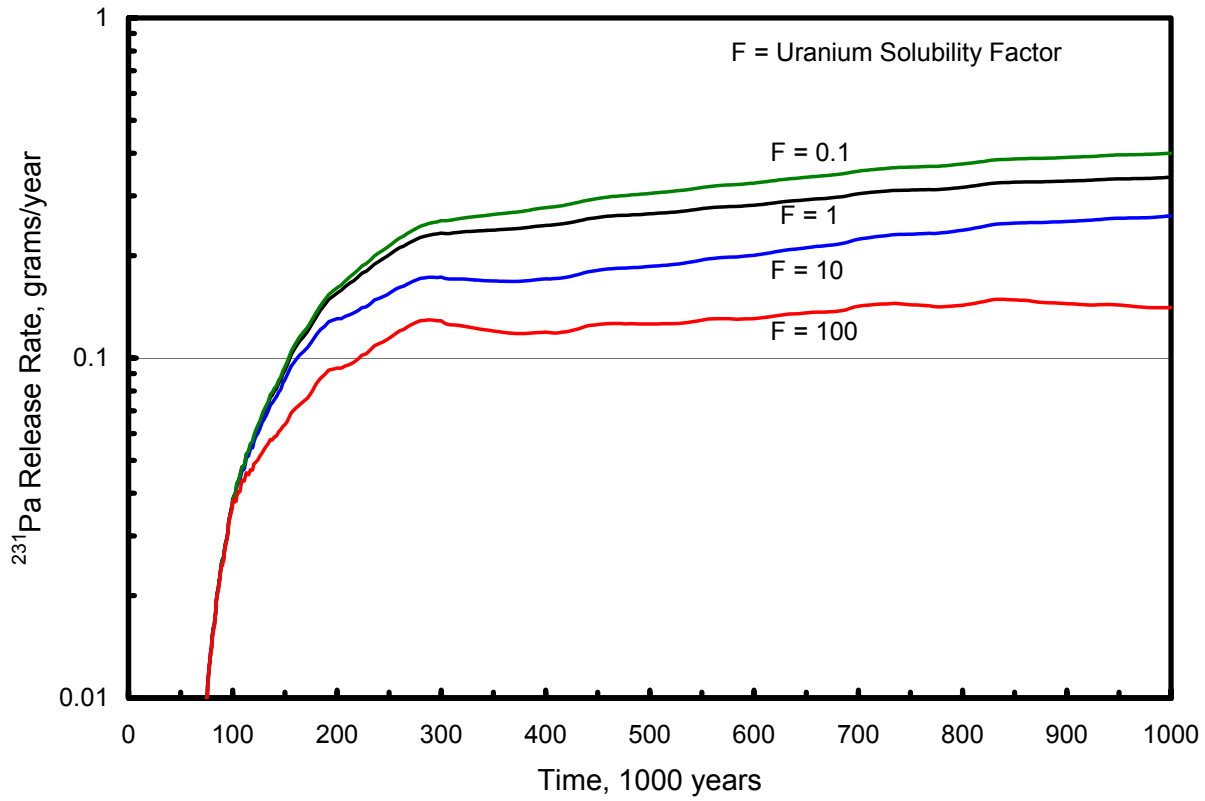

Fig. 4. Release rate of ^{231}Pa from the EBS as a function of the factor applied to the uranium solubility.

characterized by release rates that increase and have peak values shifting toward earlier times as the uranium solubility is increased. Not surprisingly, as illustrated in Fig. 4, the daughters are characterized by release rates from the EBS that decrease with increasing uranium solubility. Faster release of uranium when solubility is increased means less decay in the EBS and hence a smaller release of daughters. Conversely, slower release means more time for decay and a larger release of daughters.

IMPACT ON DOSE RATE

When the time-dependent release rates of uranium from the EBS are compared with the release rates from a boundary at the 20-km well, they are found to be nearly identical in shape and magnitude. The largest difference is that the release rate from the 20-km boundary is delayed between several thousand and a few ten thousand years relative to the release rate from the EBS. Because the shapes are similar, a reasonable estimate of the change in the contribution of most uranium isotopes to the dose rate is that the change is in the same ratio as the change in the release from the EBS. A possible exception to this is the case of ^{235}U. ^{239}Pu is a source of ^{235}U in both the EBS and the geosphere. A reduction in the release rate of ^{235}U from the EBS may increase the importance of ^{235}U resulting from the decay of ^{239}Pu. Thus, when the uranium solubility is decreased, a conservative estimate would be that the release rate of ^{235}U from the 20-km boundary is unchanged. Conversely, when the uranium solubility is increased, the importance of ^{235}U resulting from the decay of ^{239}Pu may be diminished. A conservative estimate of the impact on the release rate of ^{235}U across the boundary at the 20-km well would be that it increases by the same fraction that the uranium release rate from the EBS increases. Since the uranium contribution to the dose rate is small, even if the dose-rate changes by the factors one would estimate from Fig. 1, the change in the total dose rate would be small.

Because the dose-rate contribution from uranium isotopes is small, if changes in the uranium solubility are to have any significant impact on the dose rate, it is likely to be through uranium decay products. In the TSPA model file used for this study, the uranium decay products making the largest contributions to the dose rate are ^{227}Ac, ^{231}Pa, and ^{229}Th. For the present study, estimates of the dose-rate contributions from these radionuclides are assumed to change in the same ratio as the EBS uranium release-rate changes. As seen earlier, the release rate from the EBS for uranium decay products decreases when the uranium solubility is increased and increases when the uranium solubility decreases. Protactinium, thorium, and actinium are strongly sorbing in the TSPA model used here.[1] Implicit in the estimation procedure is the assumption that the amount of these elements that reaches the 20-km well will depend more on uranium decay near the well than on release of these elements from the EBS. However, even if the changes in the dose-rate contribution is more nearly in proportion to the change in the release rate from the EBS as shown in Fig. 4 for ^{231}Pa, the change in the dose-rate contribution would be less than a factor of three. The change in the total dose rate would be small. Using the estimation procedures outlined above, the change in the peak dose rate when the uranium solubility changes from the smallest to the largest value is less than 20%.

The TSPA model used for this work did not include dose-rate contributions for ^{230}Th, ^{226}Ra, and ^{210}Pb. According to Fig. 4.1-19a in Ref. 3, the dose-rate contributions for these radionuclides are more important than the contributions from ^{231}Pa and ^{227}Ac. ^{230}Th, ^{226}Ra, and ^{210}Pb result from the decay of ^{234}U. Assuming that the change in the dose-rate contributions from these radionuclides is in proportion to the change in the release rate of ^{234}U from the EBS, the estimated change in the total dose rate would be considerably larger than estimated in the foregoing paragraph. The change, nevertheless lies within the range determined by the 5th and 95th percentiles in Fig. 4.1-19a.[3] Note that some verification calculations using a simplified TSPA model[4] that includes the contribution to the dose rate from ^{230}Th, ^{226}Ra, and ^{210}Pb indicates increasing or decreasing the uranium solubility by a factor of 10 results in essentially no change in the total dose rate. This indicates that the estimation procedure used here over-estimates the increases in the dose rate.

CONCLUSIONS

In the foregoing calculations, the uranium solubility was modified simultaneously in both the waste packages and in the invert supporting the waste packages. Increasing uranium solubility causes a transition from transport dominated by advection to transport dominated by diffusion. Transport in packages that always or intermittently experience dripping water is dominated by advection. Waste packages that never experience dripping water are, nevertheless, able to release radionuclides once waste package failure occurs. Transport takes place via diffusion through a thin film of water that is assumed in the TSPA model to be always present in the porosity of the degraded waste forms and in the invert. Because the model imposes a zero-concentration boundary condition at the outside of the EBS, the gradient driving the diffusive transport tends to be proportional to the solubility. The transition to diffusive transport

occurs earlier for CDSP waste packages than for CSNF waste packages because waste forms degrade more rapidly in the CDSP packages. If the zero-concentration boundary condition were changed in the TSPA model, then changes in the release rate of uranium and its daughters from the EBS may be smaller than calculated here.

During much of the first 100,000 years, increasing or decreasing the uranium solubility by a factor of about 10 or less results in a proportionate increase or decrease in the uranium release rate. Between an increase of a factor of 10 and a factor of 100, this proportionality begins to break down. At later times, for packages in always and intermittent drip environments, the release of uranium tends to be controlled by the degradation of the waste form. In this case, changes in the uranium solubility do not result in proportionate changes in the release rate. The release rate from CSNF packages in never drip environments tends to be controlled by diffusive transport for uranium solubilities up to nearly 100 times nominal. Increases in the release rate of uranium from the EBS are accompanied by decreases in the release rate of uranium decay products since they do not have time to be created before crossing the EBS boundary.

Even though the release rate of uranium from the EBS may be fairly sensitive to the uranium solubility, the estimated effect on the dose rate at the 20-km well is less sensitive. This is because uranium and its daughters make small contributions to the dose rate when compared with the contribution from ^{237}Np.

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